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Method for providing synthesis gas by means of an additional electric heater

The invention relates to a reformer for the steam reforming of a hydrocarbon-containing mixture, to a plant for ammonia synthesis, hydrogen synthesis, methanol synthesis and/or as an ammonia synthesis-urea synthesis complex, to a method for producing synthesis gas, and to the use of the reformer of the invention for producing a synthesis gas mixture.

Ammonia is the second most widely produced synthetic chemical in the world (Ullmann's Encyclopedia of Industrial Chemistry, 2012, Wiley-VCH Verlag GmbH &Co. KGaA, Weinheim, DOI:10.1002/14356007.o02_o11, hereinafter "Ullmann's").

This ammonia is produced essentially from the elements hydrogen and nitrogen and an iron catalyst. The temperatures vary frequently in the range between 400°C and 500°C under a pressure over 100 bar. The key factor for the operating costs lies in the provision of hydrogen from synthesis gas production (Ullmann's, page 139).

Ammonia is preferably generated, accordingly, in the basic way described, for example, in Holleman, Wiberg, Lehrbuch der Anorganischen Chemie, 102 edition, 2007, pages 662-665 (ISBN 978-3-11-017770-1), based on the "Haber-Bosch process", from the elements in accordance with equation [1]:

$$3\ H_2 + N_2 \rightleftharpoons 2\ NH_3 + 92.28\ kJ \qquad [1]$$

The reactant nitrogen (N_2) may be obtained, for example, by low-temperature air separation or by reduction of oxygen in air by combustion. The hydrogen is obtained preferably via a "steam reforming process" in accordance with equation [2]:

$$C_nH_2m + nH_2O \rightleftharpoons (n+m)\ H_2 + nCO \qquad [2]$$

In the subsequent "carbon dioxide conversion" there is a further reaction in accordance with equation [3]:

$$CO + H_2O \rightleftharpoons CO_2 + H_2 \qquad [3]$$

The carbon dioxide (CO_2) formed in accordance with equation [3] serves preferably as a carbon dioxide source for the urea synthesis in accordance with equations [4] and [5].

$$2\,NH_3 + CO_2 \rightleftharpoons H_2N\text{-}COONH_4 \qquad [4]$$

$$H_2N\text{-}COONH_4 \rightleftharpoons (NH_2)_2CO + H_2O \qquad [5]$$

In the primary reformer, in the endothermic reforming reaction, methane is split by means of steam into hydrogen and carbon monoxide (and also, in part, CO_2). In the prior art it is customary for the necessary energy for heating the catalyst/gas/steam mixture to be accomplished exclusively by way of the reformer burners. The reformer burners, through the combustion of the air/natural gas mixture, transfer the heat by means of thermal radiation to the outer walls of the reformer tubes. These subsequently transfer the heat by means of thermal conduction to the catalyst and subsequently by convection to the gas/steam mixture. Considerable disadvantages of the system are the compromises between the maximum possible introduction of heat (wear driver) and the necessary introduction of heat. Inadequate introduction of heat has the effect that some of the water/natural gas mixture does not undergo catalytic reaction, referred to as “methane slip”.

In practice, however, the plant operator is always concerned to have the reformer burners running at full load in order to produce as much synthesis gas as possible. This operation in turn can considerably reduce the lifetime of the reformer tubes. Moreover, the catalyst volume of a reformer tube is limited by the maximum possible internal tube diameter of the reformer tubes and also by the nature and quantity of the unilateral introduction of heat. A large tube diameter makes it possible to accommodate more catalyst per reformer tube, but in that case the heat that is present no longer entirely reaches the internal catalyst near to the tube axis.

The natural gas, moreover, is used for twin purposes. It serves on the one hand as heating gas for the endothermic steam reforming process and also, on the other hand, as a supplier of hydrogen for the synthesis gas production. In the long term this is problematic, since fossil fuels are finite and are associated with rising costs. A complicating factor is that when the primary reformer is fired with natural gas, CO_2 and nitrogen oxides are emitted. Future emissions limits may then become a problem for chemical plants unless the plants possess dedicated elements for air cleaning, which is very rarely the case. This problem area is therefore associated with additional capital costs and operating costs.

EP 1 055 637 A1 discloses a method for producing a synthesis gas stream rich in carbon monoxide and hydrogen, where a catalyst in a porous support structure is used.

EP 3 075 705 A1 discloses a method and a plant for steam reforming, wherein at least some of the feedstock stream outside the first steam reformer is heated at least partly using electrical energy.

EP 0 830 893 A1 discloses a catalyst charge for the reforming of hydrocarbon-containing vapor mixtures.

DE 10 2015 013 071 A1 discloses a furnace for the steam reforming of a hydrocarbon-containing feedstock stream, where an inductor for inductive heating is mounted on a tube section.

The object of the present invention is to provide a reformer which can be heated flexibly as a function of the changing energy supply and which at the same time exposes the reformer components to as little physical loading as possible.

The object of the invention is achieved, surprisingly, by a reformer for the steam reforming of a hydrocarbon-containing mixture, as claimed in claim 1. Further advantageous embodiments are found in the dependent claims.

The invention additionally embraces a plant for ammonia synthesis, hydrogen synthesis, methanol synthesis and/or as an ammonia synthesis-urea synthesis complex.

The invention additionally embraces a method for producing synthesis gas. Further advantageous embodiments are found in the respective dependent claims.

The invention additionally embraces the use of the plant of the invention for producing a synthesis gas mixture in ammonia synthesis.

The reformer of the invention for the steam reforming of a hydrocarbon-containing mixture comprises at least the components below.

The fundamental construction of a reformer for providing hydrogen, preferably a primary reformer and a secondary reformer and/or an autothermal reformer, is known to the skilled person. The formation of hydrogen in this case takes place fundamentally according to the above equation [2]

$$C_nH_2m + nH_2O \rightleftharpoons (n+m)\ H_2 + nCO \qquad [2]$$

A depiction of the functioning of the reformer is found for example in Ullmann's, chapter 6.1.1, pages 174 to 179. The reformer comprises a combustion chamber and at least one burner disposed within the combustion chamber. A first reactor tube is disposed at least sectionally within combustion chambers. The expression "at least sectionally" means in the sense of the invention that at least part or the whole part of the first reactor tube runs within the combustion chamber. In general only a partial section of the first reactor tube passes through the combustion chamber; in other words, the first reactor tube is led into the combustion chamber and then led out of the combustion chamber. Disposed within the first reactor tube is a catalyst, based for example on nickel oxide on a support material. The reformer of the invention is characterized in that an electrically heatable heating element is disposed within the first reactor tube. The reformer tubes are under an inner overpressure. Since the tensile strength of steel decreases sharply as the outer wall temperature of the tube goes up, there is an upper limit on the density of heat flow on the outer and inner wall of the tube. If this upper limit is exceeded, there is a rapid drop in the lifetime of the tube. By virtue of the additional inner electrically heatable heating element in accordance with the invention, the restriction governing the inner wall and outer wall diameters can be eliminated; the status today is limited by the maximum possible introduction of heat and must always be designed so as to take account of pressure/temperature/wear. The invention therefore enables an improved temperature profile over the cross section of the catalyst bed. The catalyst, then, is utilized more effectively and so reduces the methane slip with a lower outer thermal load on the reformer tubes.

Preferably, additionally, there is a second reactor tube with a catalyst disposed within the second reactor tube, without an electrically heatable heating element as described above. The invention therefore also embraces the combination of first reactor tubes with electrical heating elements and second reactor tubes without electrical heating elements.

In one preferred embodiment, the first reformer tube, in the region of the electrically heatable heating element, has protrusions or constrictions. These protrusions or constrictions enable an individually adjustable heating of individual sections of the first reformer tubes.

The electrically heatable heating element preferably comprises an inductively heatable heating element. The expression "inductively heatable" in the sense of the invention preferably embraces "the heating of electrically conductive materials by induced high-frequency currents within the material" (Dictionary of Science and Technology, edt. Christopher Morres, 1992, p. 1101, ISBN: 0-12-200400-0). More preferably the inductively heatable heating element has at least one heating tube (for example containing ceramics, glass and/or metals) and, wound at least sectionally around the heating tube (6), a metallic wire and/or a metallic coating (7b), more preferably in the form of coillike conductor tracks. The metallic wire and/or the metallic coating are preferably connected to an electrical alternating current source.

The heating tube preferably has an at least sectionally metallic surface. The metallic surface improves the thermal conduction from the heating tube into the catalyst bed of the first reactor tube.

With more particular preference, the metallic (electrically conductive) wire lies on the heating tube via notches. The notches improve the fastening, “adhesion” of the metallic wire on the heating tube.

The ratio of diameter of the first reactor tube, $\varnothing_{2a}$, to the diameter of the heating tube, $\varnothing_{6}$, expressed as $\varnothing_{2a}/\varnothing_{6}$, is preferably from 100 to 2, more preferably from 10 to 5.

In one preferred embodiment, the metallic wire and/or the metallic surface and/or the metallic coating comprises iron, cobalt, nickel, copper, silver, chromium, and also semimetals such as graphite, silicon and ceramic coatings.

The metallic wire and/or the metallic coating are preferably connected to an electrical alternating current source.

The electrical heating element ~~preferably~~ comprises an electrical resistance heating element. This element preferably comprises an ohmic resistor and may be configured for example in the form of a metal wire or metal plate. The electrical resistor may be connected to an alternating current source and to a direct current source. The heat that is liberated when current flows through the electrical resistor is utilized in order to heat the electrical resistance heating element and also the surrounded catalyst bed. By virtue of the additional inner electrically heatable heating element in accordance with the invention, the restriction governing the inner wall and outer wall diameters can be eliminated; the status today is limited by the maximum possible introduction of heat and must always be designed so as to take account of pressure/temperature/wear. The invention therefore enables an improved temperature profile over the cross section of the catalyst bed. The catalyst, then, is utilized more effectively and so reduces the methane slip with a lower outer thermal load on the reformer tubes.

More preferably the electrical resistance heating element comprises an electrical heating resistor and a heating jacket. The electrical heating resistor preferably comprises metallic conductors. The heat in this case is delivered in line with the material-specific resistance. The electrical heating resistor, in the form for example of a metal plate or a metal-coated ceramic plate, is surrounded by a heating jacket. The heating jacket, comprising – for example - suitable ceramic materials, prevents direct contact with the catalyst bed and the occurrence of “hotspots” on the catalyst. The expression “suitable ceramic materials” preferably embraces ceramic materials or material mixtures with ceramic fractions that are known to the skilled person and that are stable or exhibit only little physical wear at the operating temperatures of the reformer. Illustrative ceramics may contain Al_2O_3, MgO, ZrO_2, aluminum titanates, silicides, carbides, nitrides, etc.

The heating jacket preferably comprises electrical insulators, preferably oxide ceramics and/or nonoxide ceramics.

In a further embodiment, the electrical resistance heating element (5b) comprises a first electrical heating resistor and a second heating resistor, more preferably further heating resistors. The first electrical heating resistor and the second electrical heating resistor may have different dimensions. This includes, for example, different lengths and diameters, or else the arrangement of the electrical heating resistors in the electrical resistance heating element. By way of this arrangement it is possible to achieve different and adaptable heating powers on the part of the electrical resistance heating element.

The catalyst preferably comprises a ferromagnetic catalyst, preferably iron, cobalt, nickel and/or compounds and/or mixtures thereof. Using a ferromagnetic catalyst makes it possible to carry out direct heating and hence enables better temperature distribution of the catalyst in the heating tube and in the electromagnetic induction field generated therein.

The invention further embraces a plant for ammonia synthesis, hydrogen synthesis, methanol synthesis and/or as an ammonia synthesis-urea synthesis complex, comprising a reformer of the invention as described above.

The invention further embraces a method for producing synthesis gas, at least comprising the following steps: in a first step a), a hydrocarbon-containing starting mixture, more preferably natural gas, and steam are provided. In a subsequent step b), a reformer as described above is charged with the provided hydrocarbon-containing starting mixture and steam. The fundamental operating conditions are found for example in Ullmann’s, chapter 6.1.1, pages 174 to 179. Thereafter, in step c), a synthesis gas mixture is obtained, preferably a synthesis gas mixture comprising the components according to equation [2]. The method is suitable for use in plants for ammonia synthesis, hydrogen synthesis, methanol synthesis, and in ammonia-urea synthesis complexes (i.e., combined plants for the synthesis both of ammonia and optionally carbon dioxide and also for the synthesis of urea from ammonia and carbon dioxide).

The charging in step b) takes place preferably at a temperature of 300°C to 700°C under a pressure of 20 bar to 50 bar.

The invention further embraces the use of the reformer of the invention for producing a synthesis gas mixture.

Additionally, the invention is elucidated in more detail by means of the following figures: these figures do not limit the scope of protection of the invention, instead serving only for illustrative elucidation. The figures are not to scale.

Figure 1 shows a schematic drawing of the reformer of the invention for the steam reforming of a hydrocarbon-containing mixture,

figure 2 shows a schematic drawing of the first reactor tube of the invention,

figure 3 shows an enlarged detail of the first reactor tube,

figure 4 shows an enlarged detail of the electrical resistance heating element,

figure 5 shows a schematic drawing of a further embodiment of the first reactor tube of the invention,

figure 6 shows an enlarged detail of a preferred electrical resistance heating element, and

figure 7 shows a schematic drawing of a preferred first reactor tube.

Figure 1 shows a schematic drawing of the reformer of the invention for the steam reforming of a hydrocarbon-containing mixture. The reformer comprises a combustion chamber (1) and burners (4) disposed within the combustion chamber. A first reactor tube (2a) is disposed at least sectionally within the combustion chamber (1). The expression “at least sectionally” means in the sense of the invention that at least a part or the whole part of the first reactor tube runs within the combustion chamber (1). Disposed within the first reactor tube (2a) is a catalyst (3), not shown. Also disposed within the first reactor tube (2a) is an electrically inductively heatable heating element (5a). In addition, furthermore, there may be second reactor tubes (2b) with catalyst (3), not shown, which for reasons of cost or for temperature control, for example, may include no inductively heatable heating element (5a).

Figure 2 shows a schematic drawing of the first reactor tube (2a) of the invention. Located within the first reactor tube (2a) is a catalyst (3) and an inductively heatable heating element (5a). The inductively heatable heating element (5a) comprises a heating tube (6), a metallic wire (7a) wound at least sectionally around the heating tube (6) like a coil (or metallic coating (7b)) and may likewise be filled with the catalyst (3). In principle, the catalysts (3) within the inductively heatable heating element (5a) and within the space between the inductively heatable heating element (5a) and the interior of the first reactor tube (2a) may be identical or else different. The metallic wire (7a) wound like a coil around the heating tube (6) is connected to an alternating current source, not shown. The heating of the heating tube (6) via the wound metallic wire (7a) allows the first reactor tube (2a) to be heated from the inside and therefore lowers the physical stresses through the simultaneous introduction of heat on the part of the burners (4). A hydrocarbon-containing starting mixture (9) and steam (10) enter the first reactor tube (2a) and depart the first reactor tube (2a) as a synthesis gas mixture (11).

Figure 3 shows an enlarged detail of the first reactor tube (2a). The heating tube (6) is filled (optionally) with catalyst (3) and at least sectionally, for better thermal conduction, has a (thermally conducting) metallic coating (7c). The metallic wire (7a) wound like a coil around the heating tube (6) is connected to an alternating current source, not shown. Notches (8) in the heating tube (6) improve the hold of the metallic wire (7a) on the surface of the heating tube, and additionally this creates a planar surface (8a), which allows improved transfer of heat by means of thermal conduction.

Figure 4 shows an enlarged detail of the electrical resistance heating element (5b). The electrical resistance heating element (5b) comprises an electrical heating resistor (13) and a heating jacket (12). The electrical heating resistor (13) may be electrically connected to a direct current source (indicated by + -) or an alternating current source, not shown.

Figure 5 shows a schematic drawing of a further embodiment of the first reactor tube (2a) of the invention. Disposed within the first reactor tube (2a) is a catalyst (3). The electrical resistance heating element (5b) shown in figure 4 is disposed within the catalyst (3) and enables a more uniform heating of the catalyst (3) and reduces the physical stresses occurring within the first reactor tube (2a). The electrical resistance heating element (5b) may be configured, as shown in figure 5, as separate elements in each case or else as a connected totality, connected in series or in parallel, of respectively individual electrical resistance heating elements (5b1 + 5b2 + 5b3 + …).

Figure 6 shows an enlarged detail of a preferred electrical resistance heating element (5b). The electrical resistance heating element (5b) comprises a first electrical heating resistor (13a), a second electrical heating resistor (13b) and a heating jacket (12). The electrical heating resistors (13a/13b) may be electrically connected to a direct current source (indicated by + -) or to an alternating current source, not shown. The electrical resistance element (5b) may also comprise a plurality of first and second heating resistors (13a/13b). This is indicated schematically in the circle. The first electrical heating resistor (13a) and the second heating resistor (13b) may have different dimensions. This includes, for example, different lengths and diameters or else the arrangement of the electrical heating resistors (13a/13b) in the electrical resistance heating element (5b). By way of this arrangement it is possible to achieve different and adaptable heating powers on the part of the electrical resistance heating element (5b).

Figure 7 shows a schematic drawing of a preferred first reactor tube (2a). The electrical resistance element (5b) comprises a plurality of first and second heating resistors (13a/13b). These heating resistors (13a/13b) are preferably different in their dimensions in terms of the heating power. The electrical resistance element (5b) is disposed in the first reactor tube (2a) via an electrical contacting/mechanical securement (14). The first reactor (2a) has a protrusion (15) in the region of the electrical resistance element (5b). These protrusions (15) or else constrictions enable an individually adjustable heating of individual sections of the first reformer tubes (2a).

List of reference symbols

(1)	combustion chamber
(2a)	first reactor tube
(2b)	second reactor tube
(3)	catalyst
(4)	burner
(5)	electrically heatable heating element
(5a)	inductively heatable heating element
(5b)	electrical resistance heating element
(6)	heating tube
(7a)	metallic wire
(7b)	metallic coating
(8)	notches
(8a)	contact face
(9)	hydrocarbon-containing starting mixture
(10)	steam
(11)	synthesis gas mixture
(12)	heating jacket
(13)	electrical heating resistor
(13a)	first electrical heating resistor
(13b)	second electrical heating resistor
(13i)	further i-th heating resistor with i greater/equal to 3 and i (integer) = 3, 4, 5, 6, 7, …
(14)	electrical contacting/mechanical securement
(15)	protrusion/constriction of the first reactor tube

Patentkrav

1. Reformer til dampfreformering af en carbonhydridholdig blanding, der mindst omfatter:
a.) et brændkammer (1),
b.) en brænder (4), der er anbragt i brændkammeret (1);
c.) et første reaktorrør (2a), der i det mindste afsnitsvist er anbragt i brændkammeret (1);
d.) en katalysator (3), der er anbragt i det første reaktorrør (2a);
kendetegnet ved, at et varmeelement (5), der kan opvarmes elektrisk, er anbragt i det første reaktorrør (2a), at varmeelementet (5), der kan opvarmes elektrisk, strækker sig i brændkammeret (1), hvor det elektriske varmeelement (5) omfatter et elektrisk modstandsvarmeelement (5b).

2. Reformer ifølge krav 1, **kendetegnet ved, at** et andet reaktorrør (2b) med en katalysator (3), der er anbragt i det andet reaktorrør (2b), uden et varmeelement (5), der kan opvarmes elektrisk, i det andet reaktorrør (2b) i det mindste afsnitsvist er anbragt i brændkammeret (1).

3. Reformer ifølge krav 1 eller 2, **kendetegnet ved, at** det første reformerrør i området af varmeelementet (5), der kan opvarmes elektrisk, omfatter udbulinger eller indsnævringer.

4. Reformer ifølge et af kravene 1 til 3, **kendetegnet ved, at** varmeelementet (5), der kan opvarmes elektrisk, omfatter et varmeelement (5a), der kan opvarmes induktivt.

5. Reformer ifølge krav 4, **kendetegnet ved, at** varmeelementet (5a), der kan opvarmes induktivt, omfatter mindst et varmerør (6) og en metallisk tråd (7a), der i det mindste afsnitsvist er viklet omkring varmerøret (6), eller en metallisk coating (7b), fortrinsvis i form af spoleformede lederbaner.

6. Reformer ifølge krav 5, **kendetegnet ved, at** varmerøret (6) har en i det mindste afsnitsvist metallisk overflade (7c).

7. Reformer ifølge krav 5 eller 6, **kendetegnet ved, at** den metalliske, elektrisk ledende tråd (7a) ligger an mod varmerøret (6) via indsnit (8).

8. Reformer ifølge et af kravene 5 til 7, **kendetegnet ved, at** varmerøret (6) er fyldt med katalysatoren (3) eller en anden katalysator, der adskiller sig fra katalysatoren (3).

9. Reformer ifølge et af kravene 5 til 8, **kendetegnet ved, at** katalysatoren (3) er anbragt i det første reaktorrør (2a) og i varmerøret (6).

10. Reformer ifølge et af kravene 5 til 9, **kendetegnet ved, at** forholdet mellem det første reaktorrørs (2a) $Ø_{2a}$ diameter og varmerørets (6) $Ø_6$ diameter udtrykt som $Ø_{2a}/Ø_6$ ligger ved 100 til 2, fortrinsvis 10 til 5.

11. Reformer ifølge et af kravene 5 til 10, **kendetegnet ved, at** den metalliske tråd (7a) og/eller den metalliske overflade (7c) og/eller den metalliske coating (7b) omfatter jern, kobalt, nikkel, kobber, sølv, krom.

12. Reformer ifølge et af kravene 5 til 11, **kendetegnet ved, at** den metalliske tråd (7a) og/eller den metalliske coating (7b) er forbundet med en elektrisk vekselstrømkilde.

13. Reformer ifølge krav 12, **kendetegnet ved, at** det elektriske modstandsvarmeelement (5b) omfatter en elektrisk varmemodstand (13) og en varmekappe (12).

14. Reformer ifølge krav 13, **kendetegnet ved, at** varmekappen (12) omfatter elektriske isolatorer, fortrinsvis oxidkeramikker og/eller ikke-oxidkeramikker.

15. Reformer ifølge et af kravene 12 til 14, **kendetegnet ved, at** det elektriske modstandsvarmeelement (5b) omfatter en første elektrisk varmemodstand

(13a) og en anden varmemodstand (13b), fortrinsvis yderligere varmemodstande (13i).

16. Reformer ifølge et af kravene 1 til 15, **kendetegnet ved, at** katalysatoren (3) omfatter en ferromagnetisk katalysator, fortrinsvis jern, kobalt, nikkel og/eller forbindelser og/eller blandinger deraf.

17. Anlæg til ammoniaksyntese, hydrogensyntese, metanolsyntese og/eller som ammoniaksyntese-urinstofsyntesekompleks omfattende en reformer ifølge et af kravene 1 til 16.

18. Fremgangsmåde til fremstilling af syntesegas, mindst omfattende trinnene:
a.) at tilvejebringe en carbonhydridholdig udgangsblanding (9) og vanddamp (10)
b.) at påvirke en reformer ifølge et eller flere af kravene 1 til 16 med den tilvejebragte carbonhydridholdige udgangsblanding (9) og vanddamp (10); og
c.) at opnå en syntesegasblanding (11).

19. Fremgangsmåde ifølge krav 18, **kendetegnet ved, at** påvirkningen sker i trin b) ved en temperatur på 300° C til 700° C og et tryk på 20 bar til 50 bar.

20. Anvendelse af en reformer ifølge et af kravene 1 til 16 til fremstilling af en syntesegasblanding.

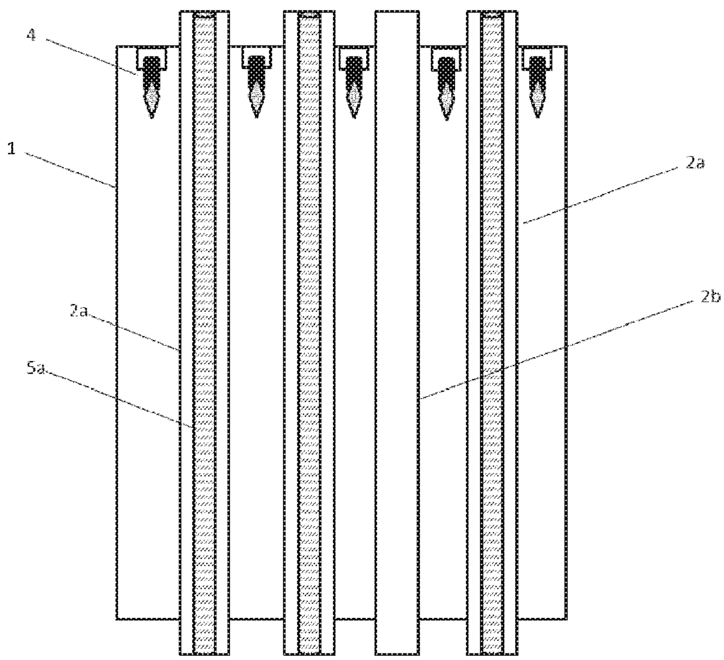

Figure 1

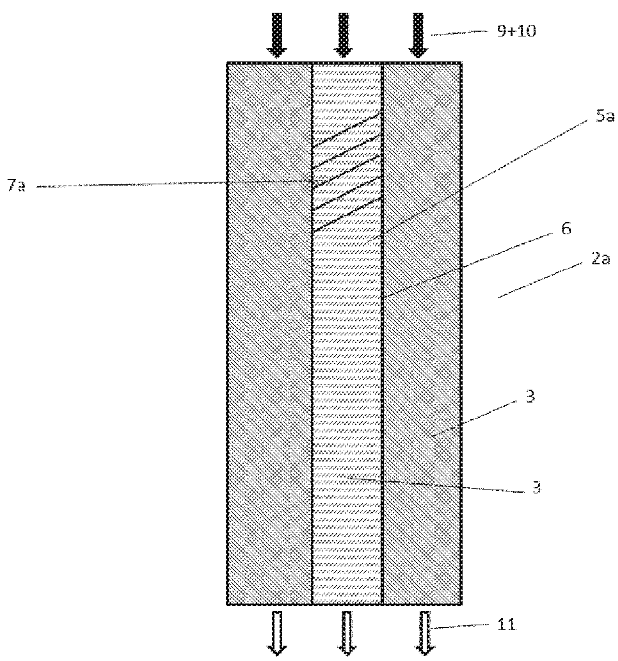

Figure 2

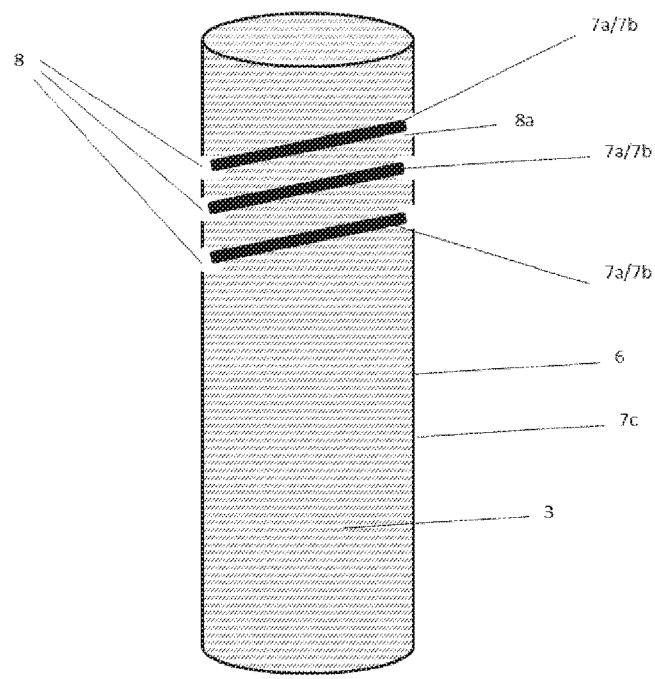

Figure 3

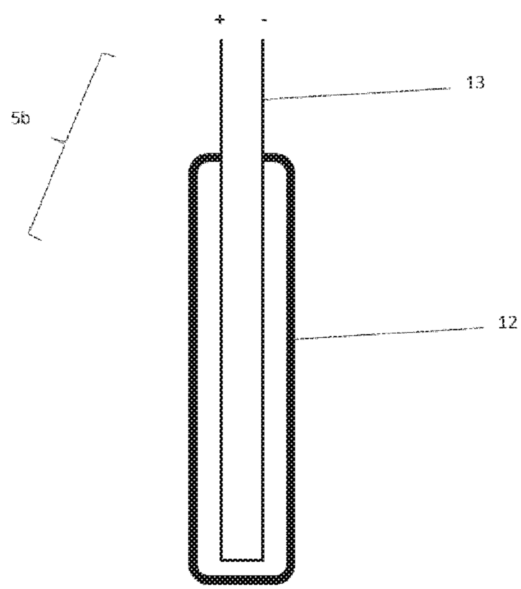

Figure 4

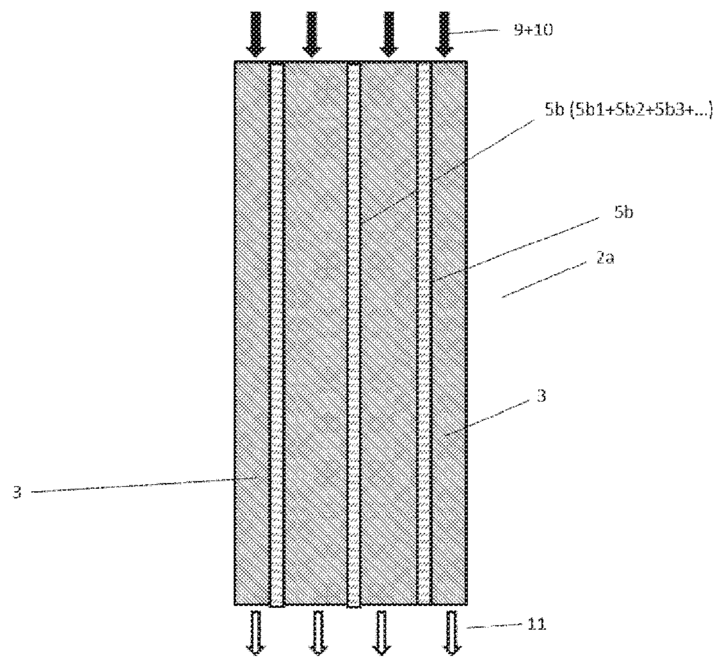

Figure 5

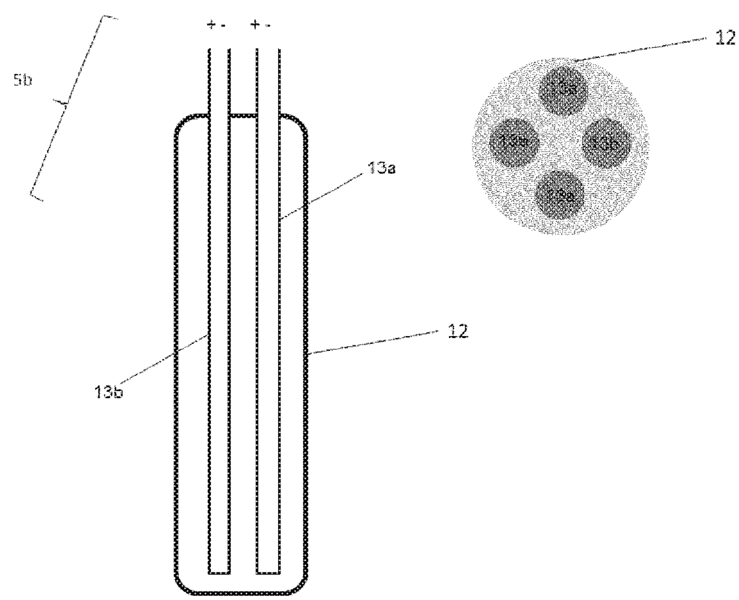

Figure 6

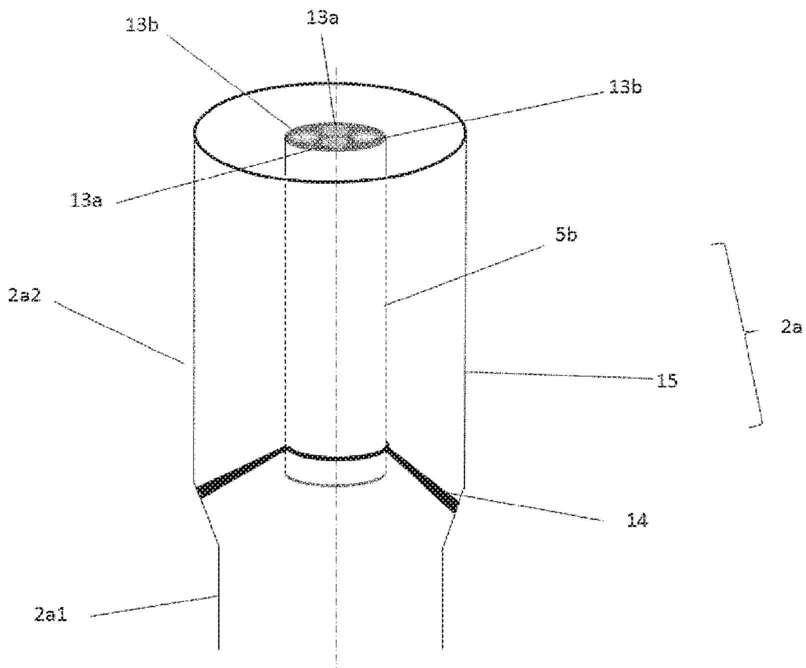

Figure 7